The lateral growth of GaN by sublimation method

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We have applied sublimation method for laterally growing GaN. The principal conclusions are that the bulk growth of GaN can occur significantly for the temperature of 1080ºC, reacting gas consisted with NH_3 and N_2, and substrate of metalorganic chemical vapor deposited GaN film/sapphire. The x-ray diffraction and scanning electron microscopes show the GaN grows laterally with a preferential (0002) orientation with a shape of hexagonal crystallites in a thickness of 20 μm parallel with sapphire substrate. While the photoluminescence spectra do vary, for the GaN we observe several peaks at 1.88, 2.25, and 3.40 eV and that the photoluminescence width is narrowed as the sublimation temperature increases between 990 and 1080ºC.

INTRODUCTION

GaN is one of the most promising materials for optoelectronic devices. GaN has been successfully grown by metalorganic chemical vapor deposition (MOCVD) [1] and molecular beam epitaxial (MBE) techniques on a sapphire substrate. However, heteroepitaxial growth still remains as problems in getting high quality GaN, because it is limited with a high density of dislocation and strain due to a large difference in lattice constants and thermal expansion coefficients between hetero-epilayer and substrate [2]. Thus the best GaN may be prepared with homoepitaxial growth on bulk GaN [3].

A sublimation technique has been suggested for the thick homoepitaxial growth of GaN and investigated by Satoshi Kurai, etc. [2,3]. The authors reported they obtained continuous crystallites on the MOCVD-GaN by the sublimation method rather than other substrates such as sapphire, scratched sapphire, SiO_2, and Si.

In contrast to other growing techniques, sublimation method is relatively easy, fast and economic. However, the method has some difficulty in growing of bulk GaN. For example, at higher temperatures evaporization of GaN is congruent only under Langmuir conditions [4], but higher pressures evaporation of GaN becomes non-congruent and liquid Ga remains on the GaN surface. There remain nitrogen molecules in the vapor phase more than GaN molecules and lack of chemical reactivity of the nitrogen molecules on the GaN surface. Hence we use the flow of ammonia and compensate the decomposition for the process of GaN growth [5]. With this motivation we studied the sublimation technique for the bulk GaN growth and investigated the structure corresponding to structural and optical properties with scanning electron microscopes (SEM), x-ray diffraction (XRD), and photoluminescence (PL).

SAMPLES AND MEASUREMENT DETAILS

Two different types of substrate have been used in the experiments. The first type consisted of sapphire (0001), while the second type of MOCVD-GaN/sapphire (0001); the second type was fabricated in the sequence of sapphire (0001)/metal organic chemical vapor deposited GaN film. We used commercially available GaN powder (electronic grade, purity: 4N). Figure 1 shows a schematic diagram for the sublimation system of GaN growth [3]. A graphite reactor is inserted in a fused-quartz tube and resistively heated to 1150ºC. NH_3 and N_2 are introduced into the reactor through flow-controlled-meters to 100 sccm. Present sublimation system is controlled with a pressure of 1×10^{-6} Torr through 3 atm. We examined sublimation of GaN with the combined gas pressure of NH_3 and N_2 from 1×10^{-3} to 1 atm and the growth duration of 0.5 to 4 hours. The gap between powder and substrate is controlled 0 to 2 cm. With an hour of sublimation at about 1050ºC and 0.5 atm GaN grows with the thickness of about 20 μm.

EXPERIMENTAL RESULTS AND DISCUSSION

We first present results for the surface of bulk-GaN/sapphire and bulk-GaN/MOCVD-GaN/sapphire structures. The bulk GaN crystal was produced through a sublimation process on the two types of substrate explained above. In Figure 2 we show scanning electron microscopes measured for the bulk-GaN/MOCVD-GaN/sapphire structure produced by varying sublimation temperature from 990 to 1080ºC. The surface of the structure appears with more enhanced crystallization as increased temperature. Especially, as positions of substrate and powder are switched each other, the surface becomes more continuous and crystallization is improved (i.e., powder at an upper side and substrate at a lower side). We also observe laterally grown GaN crystallites in a pyramid shape from the cross-sectional micrographs.

We next present photoluminescence (PL) spectra measured for the bulk-GaN/MOCVD-GaN/sapphire structure by varying temperature. In Figure 3 the spectrum shows largely three photoluminescence peaked near 1.88, 2.25, 3.40 eV which are explained with deep impurity levels [6-10] and interband transition, respectively. The spectra due to the deep levels are investigated at the moment and the interband spectrum is plotted with sublimation temperature. The inset in Fig. 3 shows the FWHM is narrowed with increasing the temperature.

In Fig. 4 we illustrate the x-ray diffraction pattern to investigate the crystallinity of GaN crystals. It confirms the crystals in a hexagonal structure and that the crystals grow laterally with a preferential (0002) orientation parallel with sapphire substrate.

CONCLUSIONS

We have grown bulk GaN by a sublimation technique on the substrate of sapphire and MOCVD-GaN/sapphire. The technique is efficient to grow thick 20 μm and large size GaN of 1cm×1cm in an hour. SEM, XRD, and PL are used to examine structural and optical properties of the grown bulk GaN. PL spectrum shows the measurement of FWHM is useful to investigate quality of the grown GaN, where the FWHM is narrowed with increasing sublimation temperature. Thus we conclude the enhanced conditions of sublimation can improve bulk GaN growth.

ACKNOWLEDGMENTs

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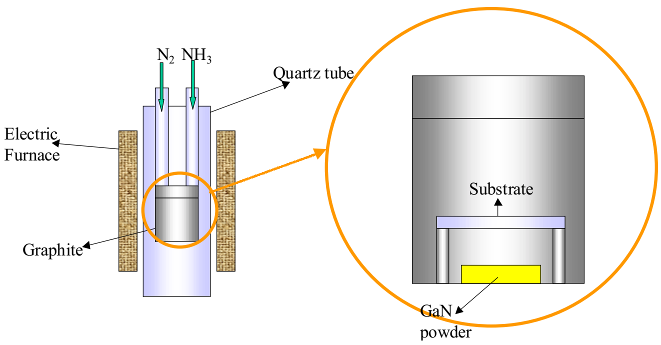

Fig. 1 Schematic diagram of the sublimation reactor for obtaining bulk GaN.

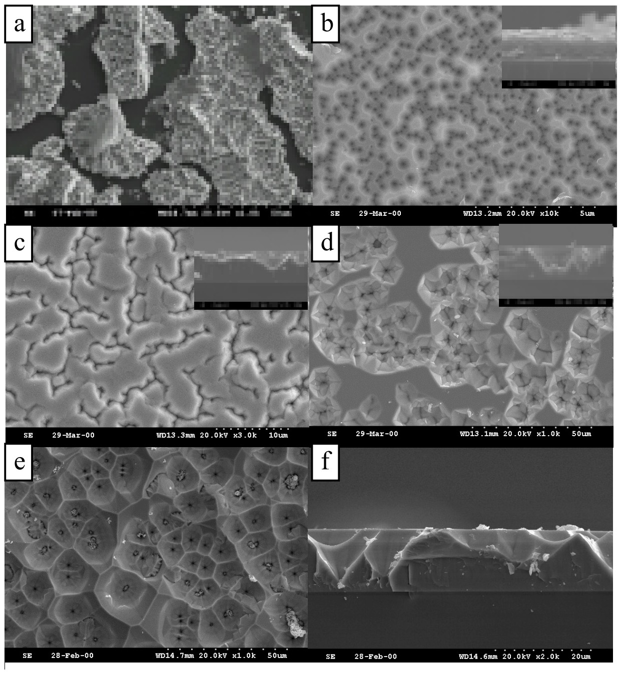

Fig. 2 (a)1080℃, (b)990℃, (c)1050℃, (d)1080℃, (e)1080℃ (swapped) : the SEM surface images for GaN.
Small (b), (c), (d) and (f) 1080℃(swapped) : the SEM cross-sectional images.
(a) bulk-GaN/sapphire. (b)–(e) bulk-GaN/MOCVD-GaN/sapphire.

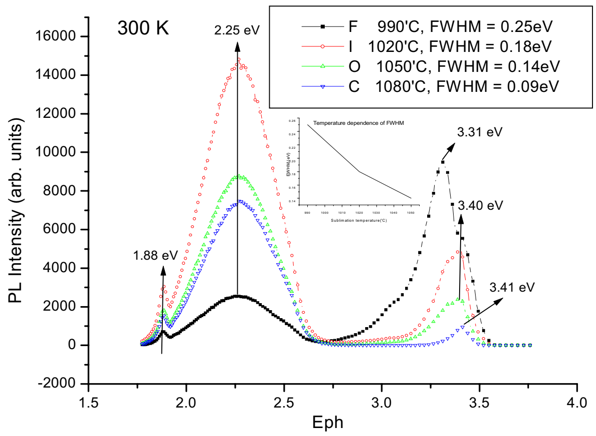

Fig. 3 PL spectra for bulk-GaN/MOCV-GaN/sapphire.

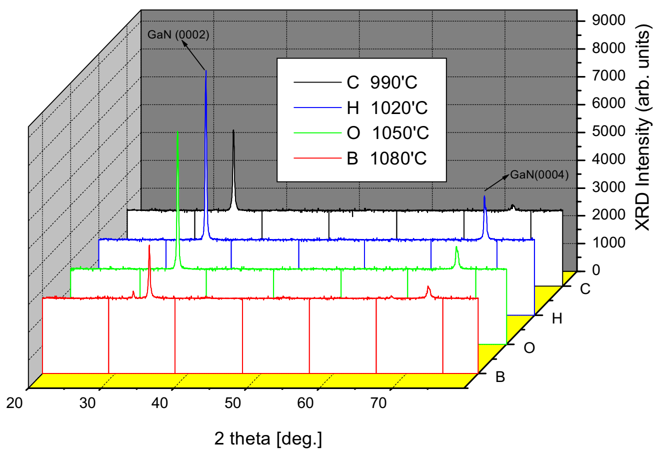

Fig. 4 XRD pattern of bulk-GaN/MOCVD-GaN/sapphire.